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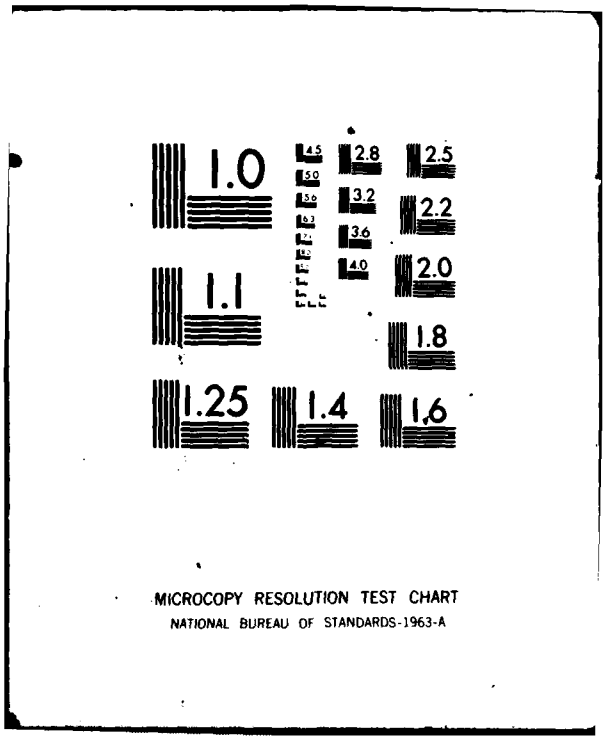
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**Abstract**

The role of energy in molecular collisions and in systems in macroscopic disequilibrium can be compactly presented and discussed using surprisal analysis. A summary of four years of progress with special reference to the dynamical foundations and the predictive apsects is presented. A list of 35 articles and books in which the work described is delineated in detail is included.

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## B. Research Objectives

Chemical reactions often release their energy in a specific fashion and thereby can be used as the pumping mechanism for lasers. Chemical reactions often consume energy in a selective fashion and therefore can be enhanced by laser pumping. A quantitative approach to these and similar observations is hampered by the great multitude of quantum states that are strongly coupled in any realistic approach. To overcome this issue and to provide an approach that can readily be used in a phenomenological fashion we have introduced the procedure often referred to nowadays as 'surprisal analysis' even though there is more to it than just the analysis. The major objectives of the last four years of research were:

1. The continued development of surprisal analysis and its application to more complex systems
2. A study of the microscopic-dynamical significance of the concept and the procedure of surprisal analysis
3. The application of information-theoretic techniques to macroscopic systems in disequilibrium.

## C. Status of Research Effort

Progress during the present project period has been such that item 1 can now be regarded as closed. All that is left are additional applications, further refinements, etc. The major objectives under item 2 have been achieved. The fundamental understanding is at hand, but practical implementations are still is the future. The opposite is the case for item 3. There have been several promising applications of surprisal analysis and several

inroads into a more fundamental understanding have been made but a unified, basic understanding is still lacking. An item by item discussion of the detailed results follows.

1. An algorithm for surprisal analysis and synthesis has been developed [5,14]. Copies of the program plus a user's manual are available for distribution and have been requested by many groups. A performance criterion[22] and a rigorous analysis of the role of experimental uncertainties, leading to error bars on the surprisal parameters [28] have been provided. A guidebook of surprisal analysis including a flow chart has been published [8]. Applications to more complex molecular processes include: unimolecular dissociations [23,29] energy transfer to polyatomic molecules [9,11], laser pumped addition reactions [32], heavy-ion collisions [16,17,19,27] and multiphoton excitation [11]. Several points previously made had to be reiterated [25,30] and a review of the first five years of surprisal analysis has been published [7].

2. The identification of constraints on dynamical grounds has been shown possible, [2,15]. The concept of a 'sum-rule', long a mainstay of surprisal synthesis has been derived as a rigorous result. This derivation is the primary practical result of these fundamental studies. Preliminary applications were to simple models of energy transfer [2] and of reactive collisions [3]. Work on more realistic potential energy surfaces is however required. Reviews of different aspects of this work have been published [13,20]. Recent work [31,35], based on earlier foundations [15] has sought to relate the

present point of view to so-called 'stochastic reduction procedures' which have recently been studied by several groups.

3. A number of practical applications (e.g. [10,11]) and several theoretical studies [21,24,31] point out that surprisal analysis has an important role to play in our understanding of systems in molecular disequilibrium. Yet, we still do not know how to identify constraints, from first principles, for such systems. The situation is particularly vexing because we do now know what to do at either of the two limiting cases: At equilibrium the constraints are the ordinary constants of the motion. For reversible evolution (e.g. isolated collisions) the constraints are the time-dependent constants of the motion [15,20]. What are they then in the intermediate situation of irreversible evolution towards equilibrium?

D. List of Publications Acknowledging AFOSR Support

1. R. D. Levine, The Information Theoretic Approach: An Interim Progress Report in ACS Symposium Series 'State-to-State Chemistry' (1977), Philip R. Brooks and Edward F. Hayes, Editors, p. 226.

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5. N. Agmon, Y. Alhassid and R. D. Levine, An Algorithm for Finding the Distribution of Maximal Entropy, J. Comput. Phys. 30, 250 (1979).

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14. N. Agmon, Y. Alhassid and R.D.Levine, An Algorithm for Determining the Lagrange Parameters in The Maximal Entropy Formalism, MIT Press, 1979, p. 207.

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17. Y. Alhassid, R. D. Levine, J. S. Karp and S. G. Steadman, Information Theoretic Analysis of Energy Disposal in Heavy Ion Transfer Reactions. Phys. Rev. C20, 1789 (1979).

18. R. D.Levine and M. Tribus, Editors, The Maximum Entropy Formalism, MIT Press, 1979, 500 p.

19. R. D. Levine and S. G. Steadman, Information Theoretic Analysis of Gamma-Ray Multiplicities in Deep Inelastic Collisions, Phys. Letts.

20. R. D.Levine, The Information Theoretic Approach to Intra-Molecular Dynamics, in Photoselective Chemistry, J. Jortner, R. D. Levine and S. A. Rice, eds., (Wiley, NY, 1980).

21. R. D. Levine, Reactions and Relaxations in Gaseous Molecular Systems - The Information Theoretic Approach (O. Laporte Memorial Lecture), in Proceedings XII Int'l. Symposium on Shock Tubes and Waves (Magnes Press, Jerusalem, 1979).

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35. R. D. Levine, Does Chaotic Time Evolution Rule Out Selective Processes in Isolated Molecules? Chem. Phys. Letts.

## E. Personnel

Y. Alhassid has participated extensively in this project as is reflected in the list of publications. He has by now submitted his work as a Ph.D. thesis, which has been judged as the 'best science Ph.D. thesis for 1980'. Dr. Alhassid is currently a Weizmann postdoctoral fellow at California Institute of Technology. R. D. Levine was awarded the Weizmann Science Prize* for his work on energy disposal in chemical reactions.

*Previous recipients include C. Pekeris (Applied Math), Y. Ne'eman (Physics), E. Katzir (Biophysics) and M. Sela (Immunology), all of whom are members of the National Academy of Science.

## F. Interactions

Reports on the research carried out under the grant were presented at international conferences, Gordon Conferences (1977, 1979) and many colloquia, seminars and summer institutes. In particular, the Arthur D. Little lectures at M.I.T. (1978) and the Otto Laporte lecture at the XII international conference on Shock Tubes and Waves (1979). A number of these presentations have been published as is reflected in the list of publications. Two international conferences on the subject of this proposal have been organized (The Maximum Entropy Formalism, 1978 and Photoselective Chemistry, 1978) and their proceedins have been published.

G. Other Statements

The list of publications includes one item which perhaps deserve a special mention: An original book 'Lasers and Chemical Change' by A. Ben-Shaul, Y. Haas, K. L. Kompa and R. D. Levine. The progress reported in this book reflects the vitality of current research in chemical dynamics. It is of special interest to note however that many of the topics discussed document the work of research groups sponsored by the Air Force Office of Scientific Research.

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20. ABSTRACT (Continue on reverse side if necessary and identify by block number)

The role of energy in molecular collisions and in systems in macroscopic disequilibrium can be compactly presented and discussed using surprisal analysis. A summary of four years of progress with special reference to the dynamical foundations and the predictive aspects is presented. A list of 35 articles and books in which the work described is delineated in detail is included.

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